PATENT COOPER	ATION TREATY
PATENT COOPER.  PCT  NOTIFICATION OF ELECTION  (PCT Rule 61.2)	
57	From the INTERNATIONAL BUREAU
PCT PCT	То:
NOTIFICATION OF FLECTION	Commissioner US Department of Commerce
4 10 11011011011011	United States Patent and Trademark
(PCT Rule 61.2)	Office, PCT
/	2011 South Clark Place Room CP2/5C24
	Arlington, VA 22202
Date of mailing (day/month/year)	ETATS-UNIS D'AMERIQUE
17 June 2002 (17.06.02)	in its capacity as elected Office
International application No.	Applicant's or agent's file reference
PCT/JP00/06951	JHTK-9-PCT
International filing date (day/month/year) 05 October 2000 (05.10.00)	Priority date (day/month/year) 07 October 1999 (07.10.99)
	07 October 1333 (07:10:33)
Applicant	
SHIBATA, Katsuji et al	
1. The designated Office is hereby notified of its election made  X in the demand filed with the International Preliminary  04 May 2001 (0)  in a notice effecting later election filed with the International Preliminary  2. The election X was  was not  made before the expiration of 19 months from the priority defined as 2.2(b).	Examining Authority on: 04.05.01) ational Bureau on:

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland

Authorized officer

Akiko KOYAMA

Telephone No.: (41-22) 338.83.38

Facsimile No.: (41-22) 740.14.35

### PATENT COOPERATION TREATY



**PCT** 



### INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	(Form PCT/ISA/2	of Transmittal of International Search Report 20) as well as, where applicable, item 5 below.
JHTK-9-PCT	ACTION	(Earliest) Priority Date (day/month/year)
International application No.	International filing date (day/month/year)	
PCT/JP 00/06951	05/10/2000	07/10/1999
Applicant		
HITACHI CHEMICAL CO., LTD	. et al.	
according to Article 18. A copy is being to  This International Search Report consists		
Basis of the report		
a With regard to the language, the	international search was carried out on the baless otherwise indicated under this item.	asis of the international application in the
the international search v	vas carried out on the basis of a translation of	the international application furnished to this
Authority (Rule 23.1(b)).  b With regard to any nucleotide at	nd/or amino acid sequence disclosed in the	international application, the international search
was carried out on the basis of the	ie sequence listing :	
·	onal application in written form. ernational application in computer readable fo	rm
· -	o this Authority in written form.	
	o this Authority in computer readble form.  beguently furnished written sequence listing	does not go beyond the disclosure in the
international application	as filed has been furnished.	
the statement that the infurnished	formation recorded in computer readable form	is identical to the written sequence listing has been
2. Certain claims were fo	und unsearchable (See Box I).	
3. Unity of invention is la	cking (see Box II).	
A NATING ADDRESS to the Atalo		
4. With regard to the <b>title</b> ,	submitted by the applicant.	
	ished by this Authority to read as follows:	
the text has been estable	islied by the Addictity to road actioners.	
5. With regard to the abstract,		
the text is approved as s	submitted by the applicant.	
the text has been estable	ished, according to Rule 38.2(b), by this Authone date of mailing of this international search r	ority as it appears in Box III. The applicant may, report, submit comments to this Authority.
	blished with the abstract is Figure No.	·
as suggested by the app	·	X None of the figures.
because the applicant fa		
	er characterizes the invention.	

Internetional Application No

A. CLASSIF IPC 7	COSJ7/00 COSC 63/00		
According to	International Patent Classification (IPC) or to both national classifi	cation and IPC	
B. FIELDS S	SEARCHED		
Minimum doo IPC 7	cumentation searched (classification system followed by classification ${\tt C08J-C08L}$	ition symbols)	
Documentati	on searched other than minimum documentation to the extent that	such documents are included in the fields sea	arched
Electronic da	ata base consulted during the international search (name of data b	pase and, where practical, search terms used)	
	PI Data, EPO-Internal		
C DOCUME	ENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication, where appropriate, of the	relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04,		1,11,23
	30 April 1997 (1997-04-30) & JP 08 325436 A (HITACHI CHEM 10 December 1996 (1996-12-10) abstract	CO LTD),	
х	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04, 30 April 1997 (1997-04-30) & JP 08 325437 A (HITACHI CHEM 10 December 1996 (1996-12-10) abstract	CO LTD),	1,11,23
		-/	
	rther documents are listed in the continuation of box C.	Patent family members are listed	d in annex.
*A* docum	categories of cited documents:  nent defining the general state of the art which is not  idered to be of particular relevance	*T* later document published after the into or priority date and not in conflict wit cited to understand the principle or to invention	heory underlying the
'E' earlier	r document but published on or after the international date	"X" document of particular relevance; the cannot be considered novel or cannot involve an inventive step when the constitution.	locument is taken alone
whic citati	th is cited to establish the publication date of another ion or other special reason (as specified) ment referring to an oral disclosure, use, exhibition or	<ul> <li>'Y' document of particular relevance; the cannot be considered to involve an id document is combined with one or rements, such combination being obvi</li> </ul>	nore other such docu-
othe	r means ment published prior to the international filing date but than the priority date claimed	in the art. *&* document member of the same pater	nt family
Date of th	e actual completion of the international search	Date of mailing of the international s	earch report
	26 July 2001	03/08/2001	
Name and	d mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2	Authorized officer	
	NL – 2280 HV Rijswijk Tel. (+31-70) 340–2040, Tx. 31 651 epo nl, Fay: (+31-70) 340–3016	Von Kuzenko, M	

2

International Application No

C.(Continua	ation) DOCUMENTS CONSIDER TO BE RELEVANT	
Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Х	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04, 30 April 1997 (1997-04-30) & JP 08 325438 A (HITACHI CHEM CO LTD), 10 December 1996 (1996-12-10) abstract	1,11,23
X	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 04, 31 March 1998 (1998-03-31) & JP 09 316445 A (HITACHI CHEM CO LTD), 9 December 1997 (1997-12-09) abstract	1,11,23
X	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 10, 31 August 1998 (1998-08-31) & JP 10 126052 A (HITACHI CHEM CO LTD), 15 May 1998 (1998-05-15) abstract	1,11,23

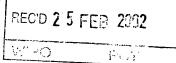
ormation on patent family members

International Application No

		Publication date	Patent family member(s)	Publication date
08325436	Α	10-12-1996	NONE	
08325437	Α	10-12-1996	NONE	
08325438	Α	10-12-1996	NONE	
09316445	Α	09-12-1997	NONE	
10126052	Α	15-05-1998	NONE	
	tent document in search report 08325436 08325437 08325438 09316445 10126052	08325436 A 08325437 A 08325438 A 09316445 A	in search report         date           08325436         A         10-12-1996           08325437         A         10-12-1996           08325438         A         10-12-1996           09316445         A         09-12-1997	in search report         date         member(s)           08325436         A         10-12-1996         NONE           08325437         A         10-12-1996         NONE           08325438         A         10-12-1996         NONE           09316445         A         09-12-1997         NONE



## PCT



### INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70) 10/088531

Applican		ent's file reference	FOR FURTHER ACTION	See Notification of Preliminary Examin	Fransmittal of International ation Report (Form PCT/IPEA/416)
Internation	onal apr	olication No.	International filing date (day/mon	th/year) Priority	/ date (day/month/year)
PCT/JF			05/10/2000	07/10	0/1999
Internation C08J7/		ent Classification (IPC) o	r national classification and IPC		
Applican HITAC		EMICAL CO., LTD.	et al.		
1. Thi	is interr d is trar	national preliminary ex nsmitted to the applica	amination report has been preparent according to Article 36.	ed by this Internation	al Preliminary Examining Authority
2. Thi	is REP	ORT consists of a tota	l of 5 sheets, including this cover	sheet.	
	been	amended and are the	nied by ANNEXES, i.e. sheets of basis for this report and/or sheets n 607 of the Administrative Instruc	containing rectificati	ons made before this Authority
The	ese an	nexes consist of a tota	I of sheets.		
					MFOR
3. Thi	is repo	rt contains indications	relating to the following items:	4	\$50 QQ 2000 PM
	Į 🗵	Basis of the report		. 🗬	
	11 <b></b>				the state of the s
ı	III 🗆	Non-establishment	of opinion with regard to novelty, i	nventive step and inc	dustrial applicability
F	v 🗆		ention		**
	v 🗵	Reasoned statemer citations and explar	nt under Article 35(2) with regard t nations suporting such statement	o novelty, inventive s	tep or industrial applicability;
\	vı 🗆	Certain documents	cited		
٧	/II 🗆	Certain defects in th	ne international application		
VI	III C	Certain observation	s on the international application		
Date of	submiss	sion of the demand	Date	of completion of this rep	ort
04/05/	2001		20.02	2002	
	ary exa	ng address of the internation	tional Author	rized officer	B SOLEON MERCAL
<u> </u>	D- Te	rropean Patent Office 80298 Munich II. +49 89 2399 - 0 Tx: 52		Kuzenko, M	(SE 2000) ST 100
	Fa	x: +49 89 2399 - 4465	Telep	hone No. +49 89 2399 8	3605





# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/JP00/06951

		is of the report	
1.	the and	receivina Office in re	ents of the international application (Replacement sheets which have been furnished to esponse to an invitation under Article 14 are referred to in this report as "originally filed" this report since they do not contain amendments (Rules 70.16 and 70.17)):
	1-36	3	as originally filed
	Clai	ms, No.:	
	1-25	5	as originally filed
2.	With lang	regard to the <b>lang</b> i uage in which the ir	uage, all the elements marked above were available or furnished to this Authority in the attendational application was filed, unless otherwise indicated under this item.
	The	se elements were a	vailable or furnished to this Authority in the following language: , which is:
		the language of a ti	anslation furnished for the purposes of the international search (under Rule 23.1(b)).
			olication of the international application (under Rule 48.3(b)).
			ranslation furnished for the purposes of international preliminary examination (under Rule
3.	With inter	n regard to any <b>nucl</b> rnational preliminary	eotide and/or amino acid sequence disclosed in the international application, the examination was carried out on the basis of the sequence listing:
		contained in the int	ernational application in written form.
		filed together with t	he international application in computer readable form.
		furnished subseque	ently to this Authority in written form.
		furnished subseque	ently to this Authority in computer readable form.
		the international ap	the subsequently furnished written sequence listing does not go beyond the disclosure in plication as filed has been furnished.
		The statement that listing has been fur	the information recorded in computer readable form is identical to the written sequence nished.
4.	The	amendments have	resulted in the cancellation of:
		the description,	pages:
		the claims,	Nos.:
		the drawings,	sheets:

5. 

This report has been established as if (some of) the amendments had not been made, since they have been

considered to go beyond the disclosure as filed (Rule 70.2(c)):





## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/JP00/06951

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

- 6. Additional observations, if necessary:
- V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- 1. Statement

Novelty (N)

Yes:

Claims

No:

Claims 1-4,11,23

Inventive step (IS)

Yes: (

Claims

No:

Claims 1-25

Industrial applicability (IA)

Yes:

Claims 1-25

No: Claims

2. Citations and explanations see separate sheet



## INTERNATIONAL PRELIMINARY

International application No. PCT/JP00/06951

**EXAMINATION REPORT - SEPARATE SHEET** 

cf V

- D1: PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04, 30 April 1997 (1997-04-30) & JP 08 325436 A (HITACHI CHEM CO LTD), 10 December 1996 (1996-12-10)
- D2: PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04, 30 April 1997 (1997-04-30) & JP 08 325437 A (HITACHI CHEM CO LTD), 10 December 1996 (1996-12-10)
- D3: PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04, 30 April 1997 (1997-04-30) & JP 08 325438 A (HITACHI CHEM CO LTD), 10 December 1996 (1996-12-10)
- D4: PATENT ABSTRACTS OF JAPAN vol. 1998, no. 04, 31 March 1998 (1998-03-31) & JP 09 316445 A (HITACHI CHEM CO LTD), 9 December 1997 (1997-12-09)
- D5: PATENT ABSTRACTS OF JAPAN vol. 1998, no. 10, 31 August 1998 (1998-08-31) & JP 10 126052 A (HITACHI CHEM CO LTD), 15 May 1998 (1998-05-15)
- 1. Documents D1 - D5 disclose liquid compositions which correspond to the liquid composition of product claim 23.
  - These liquid compositions contain amide-based solvents and alkali metal compounds.
  - In D1 to D5 the liquid composition is called "etching fluid". These fluids lead to a decomposition of the epoxy resin.
  - Therefore the teaching of these documents also foreshadows the subject-matter of method claims 1 to 4 and 11, as the epoxy resin is also decomposed. Art. 33(2) PCT.
- 2. Liquid compositions for the decomposition of cured epoxy resins containing as essential components a decomposition catalyst (alkali metal compounds) and solvents (amide based) to decompose and dissolve the epoxy resin are state of the art. Therefore the subject-matter of claims 1 to 25 does not meet the requirements of Art. 33(3) PCT.





### INTERNATIONAL PRELIMINARY **EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/JP00/06951

cf VIII

The term "decomposition catalyst" expresses only a result to be achieved without specifying relevant compounds which actually decompose under the given circumstances the epoxy resin.

It is also unclear which types of organic solvents are really to be used to "decompose" and "dissolve" the obtained decomposition-products.



### NOTIFICATION CONCERNING **SUBMISSION OR TRANSMITTAL** OF PRIORITY DOCUMENT

PCT

(PCT Administrative Instructions, Section 411)

From the INTERNATIONAL BUREAU

To:

MIYOSHI, Hidekazu 9th floor, Toranomon Daiichi Building 2-3, Toranomon 1-chome Minato-ku, Tokyo 105-0001 **JAPON** 



Date of mailing (day/month/year) 07 December 2000 (07.12.00)	MIYOSH		
Applicant's or agent's file reference  JHTK-9-PCT	IMPORTANT NOTIFICATION		
International application No. PCT/JP00/06951	International filing date (day/month/year) 05 October 2000 (05.10.00)		
International publication date (day/month/year) Not yet published	Priority date (day/month/year) 07 October 1999 (07.10.99)		
Applicant			

HITACHI CHEMICAL CO., LTD. et al

- The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).
- 2. This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.
- An asterisk(\*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.
- The letters "NR" appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, as provided by Rule 17.1(a) or (b), respectively. In such a case, the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

Priority date	Priority application No.	Country or regional Office or PCT receiving Office	Date of receipt of priority document
07 Octo 1999 (07.10.99)	11/286187	JP	28 Nove 2000 (28.11.00)
07 Octo 1999 (07.10.99)	11/286188	JP	28 Nove 2000 (28.11.00)

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland

Authorized officer

Tessadel PAMPLIEGA TOP

Facsimile No. (41-22) 740.14.35

Telephone No. (41-22) 338.83.38

### PCT

### NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

#### From the INTERNATIONAL BUREAU

To:

MIYOSHI, Hidekazu 9th floor, Toranomon Daiichi Building 2-3, Toranomon 1-chome

JAPON



Date of mailing (day/month/year)

12 April 2001 (12.04.01)

Applicant's or agent's file reference

JHTK-9-PCT

IMPORTANT NOTICE

International application No.

PCT/JP00/06951

International filing date (day/month/year)

05 October 2000 (05.10.00)

Priority date (day/month/year)

07 October 1999 (07.10.99)

**Applicant** 

HITACHI CHEMICAL CO., LTD. et al

 Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice: KR,US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:

CN, EP, SG

The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on 12 April 2001 (12.04.01) under No. WO 01/25317

#### REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a demand for international preliminary examination must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

### REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the national phase, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland

Authorized officer

J. Zahra

Telephone No. (41-22) 338.83.38 Facsimile No. (41-22) 740.14.35

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization International Bureau



### 

### (43) International Publication Date 12 April 2001 (12.04.2001)

### **PCT**

## (10) International Publication Number WO 01/25317 A3

(51) International Patent Classification?: C08J 7/00.

(21) International Application Number: PCT/JP00/06951

(22) International Filing Date: 5 October 2000 (05.10.2000)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

11/286187 7 October 1999 (07.10.1999) JP 11/286188 7 October 1999 (07.10.1999) JP

(71) Applicant (for all designated States except US): HI-TACHI CHEMICAL CO., LTD. [JP/JP]: 1-1. Nishishin-juku 2-chome, Shinjuku-ku. Tokyo 163-0449 (JP).

(72) Inventors; and

(75) Inventors/Applicants (for US only): SHIBATA, Katsuji [JP/JP]; 422-8, Okushi, Shimotsuma-shi, Ibaraki 304-0023 (JP), SHIMIZU, Hiroshi [JP/JP]; 208-1, Oaza-Nakadate,

Shimodate-shi, Ibaraki 308-0005 (JP). MATSUO, Ayako [JP/JP]: 8522-46, Yuki, Yuki-shi, Ibaraki 307-0001 (JP). HORIUCHI, Takeshi [JP/JP]: 6500-12, Tomonuma, Nogi-machi, Shimotsuga-gun, Tochigi 329-0101 (JP).

(74) Agents: MIYOSHI, Hidekazu et al.; 9th floor, Toranomon Daiichi Building. 2-3. Toranomon 1-chome, Minato-ku, Tokyo 105-0001 (JP).

(81) Designated States (national): CN. KR. SG. US.

(84) Designated States (regional): European patent (AT, BE, CH, CY, DE, DK, ES, Fl. FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

#### Published:

with international search report

(88) Date of publication of the international search report: 2 May 2002

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

A3

### (54) Title: METHOD OF TREATING EPOXY RESIN-CURED PRODUCT

(57) Abstract: The present invention is directed to a method of treating a composite material of epoxy resin-cured product and inorganic matter to easily recover the resin components in a recyclable state without causing thermal decomposition. It is also directed to a method of separating the inorganic matter. Specifically, the epoxy resin-cured product is treated with a treatment liquid which contains a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product. A composite material of epoxy resin-cured product and inorganic matter is also treated to decompose and dissolve the epoxy resin-cured product in the above-mentioned manner and the inorganic matter is then separated from the liquid.

Interr nal Application No PCT/JP 00/06951

A. CLASSIFICATION OF SUBJECT MATTER IPC 7 C08J7/00 C08L63/00

According to International Patent Classification (IPC) or to both national classification and IPC

### B. FIELDS SEARCHED

4. A.

 $\begin{array}{ccc} \text{Minimum documentation searched} & \text{(classification system followed by classification symbols)} \\ IPC & 7 & C08J & C08L \\ \end{array}$ 

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

PAJ, WPI Data, EPO-Internal

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Calegory	Challot of document, with indication. Where appropriate, of the following passages	100000000000000000000000000000000000000
χ	PATENT ABSTRACTS OF JAPAN	1,11,23
•	vol. 1997, no. 04,	, ,
	30 April 1997 (1997-04-30)	
	& JP 08 325436 A (HITACHI CHEM CO LTD),	
	10 December 1996 (1996-12-10)	
	abstract	
		1
X	PATENT ABSTRACTS OF JAPAN	1,11,23
	vol. 1997, no. 04,	
	30 April 1997 (1997-04-30)	
	& JP 08 325437 A (HITACHI CHEM CO LTD), 10 December 1996 (1996-12-10)	
	abstract	
	abstract	
	-/	
		· ·

	1/21
Special categories of cited documents:      A* document defining the general state of the art which is not considered to be of particular relevance.	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
<ul> <li>*E* earlier document but published on or after the international filling date</li> <li>*L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</li> <li>*O* document referring to an oral disclosure, use, exhibition or other means</li> <li>*P* document published prior to the international filling date but later than the priority date claimed</li> </ul>	<ul> <li>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</li> <li>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</li> <li>"&amp;" document member of the same patent family</li> </ul>
Date of the actual completion of the international search  26 July 2001	Date of mailing of the international search report 03/08/2001
Name and mailing address of the ISA  European Patent Office, P.B. 5818 Patentlaan 2  NL - 2280 HV Rijswijk  Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  Fax: (+31-70) 340-3016	Authorized officer  Von Kuzenko, M

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

Intern nal Application No PCT/JP 00/06951

A / C		PCT/JP 00/06951		
C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT  Category Citation of document, with indication, where appropriate, of the relevant passages    Relevant to claim No.				
Jaiegory °	Спанон от document, with indication, where appropriate, of the relevant passages	Relevant to claim No.		
X	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 04, 30 April 1997 (1997-04-30) & JP 08 325438 A (HITACHI CHEM CO LTD), 10 December 1996 (1996-12-10) abstract	1,11,23		
X	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 04, 31 March 1998 (1998-03-31) & JP 09 316445 A (HITACHI CHEM CO LTD), 9 December 1997 (1997-12-09) abstract	1,11,23		
X	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 10, 31 August 1998 (1998-08-31) & JP 10 126052 A (HITACHI CHEM CO LTD), 15 May 1998 (1998-05-15) abstract	1,11,23		

...ormation on patent family members

Interr nai Application No
PCT/JP 00/06951

10-12-1996	NONE	- w
10-12-1996	NONE	, The first state the corp size size due to the corp size and
10-12-1996	NONE	
09-12-1997	NONE	
15-05-1998	NONE	
	10-12-1996 09-12-1997	10-12-1996 NONE 09-12-1997 NONE

### (19) World Intellectual Property Organization International Bureau



### 

## (43) International Publication Date 12 April 2001 (12.04.2001)

### PCT

## (10) International Publication Number WO 01/25317 A2

(51) International Patent Classification<sup>7</sup>: C08J 7/00, C08L 63/00

(21) International Application Number: PCT/JP00/06951

(22) International Filing Date: 5 October 2000 (05.10.2000)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

11/286187 11/286188 7 October 1999 (07.10.1999) JP 7 October 1999 (07.10.1999) JP

(71) Applicant (for all designated States except US): HI-TACHI CHEMICAL CO., LTD. [JP/JP]; 1-1, Nishishinjuku 2-chome, Shinjuku-ku, Tokyo 163-0449 (JP).

(72) Inventors; and

(75) Inventors/Applicants (for US only): SHIBATA, Katsuji [JP/JP]; 422-8, Okushi, Shimotsuma-shi, Ibaraki 304-0023

(JP). SHIMIZU, Hiroshi [JP/JP]; 208-1, Oaza-Nakadate, Shimodate-shi, Ibaraki 308-0005 (JP). MATSUO, Ayako [JP/JP]; 8522-46, Yuki, Yuki-shi, Ibaraki 307-0001 (JP). HORIUCHI, Takeshi [JP/JP]; 6500-12, Tomonuma, Nogi-machi, Shimotsuga-gun, Tochigi 329-0101 (JP).

(74) Agents: MIYOSHI, Hidekazu et al.; 9th floor, Toranomon Daiichi Building, 2-3, Toranomon 1-chome, Minato-ku, Tokyo 105-0001 (JP).

(81) Designated States (national): CN, KR, SG, US.

(84) Designated States (regional): European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

#### Published:

 Without international search report and to be republished upon receipt of that report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: METHOD OF TREATING EPOXY RESIN-CURED PRODUCT

(57) Abstract: The present invention is directed to a method of treating a composite material of epoxy resin-cured product and inorganic matter to easily recover the resin components in a recyclable state without causing thermal decomposition. It is also directed to a method of separating the inorganic matter. Specifically, the epoxy resin-cured product is treated with a treatment liquid which contains a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product. A composite material of epoxy resin-cured product and inorganic matter is also treated to decompose and dissolve the epoxy resin-cured product in the above-mentioned manner and the inorganic matter is then separated from the liquid.

7 A2

#### DESCRIPTION

### METHOD OF TREATING EPOXY RESIN-CURED PRODUCT

### Technical Field

The present invention relates to a method of treating an epoxy resin-cured product, a method of separating a composite material of inorganic matter and epoxy resin-cured product, and a treatment liquid for an epoxy resin-cured product.

10

15

20

25

5

### Background Art

Epoxy resin-cured products are used in a variety of fields such as insulating materials, adhesives, and paints due to their superiority in electrical characteristics, heat resistance, and adhesion. However, since the epoxy resin becomes infusible and insoluble in general-purpose solvents after it is thermo-cured, it has been difficult to recycle the epoxy resin-cured products and products to which the epoxy resin-cured product adheres or on which the epoxy resin-cured product is applied. Furthermore, it has been difficult as well to separate inorganic fillers, inorganic fibers, inorganic textile fabrics, inorganic fiber nonwoven fabrics, etc., which are blended with the epoxy resin to increase the resin's mechanical properties or the like, from the resin-cured products, for recycling purpose.

A technique of grinding, pulverizing, and separating according to specific gravity is a known example of a method

of separating a printed wiring board, which is a composite material of epoxy resin-cured product and inorganic matter such as glass fiber and metal, into each component. However, although recovery and recycling of metals are possible to some extent in this method, pulverized glass fibers and resin powders thus obtained can be utilized only as fillers. Therefore their value as valuable goods is significantly low, and, in addition, it has been impossible to separate each component perfectly. Also, known examples include a method in which resins of printed wiring boards, laminates or the 10 like are thermal-decomposed to recover metals and glass fibers while the thermal-decomposed resins are gasified or liquidized for the recovery, and a method of recovering not only inorganic matter but also the thermal decomposed resins. However, these methods have several problems as follows. 15 Thermal decomposition of resins requires high temperatures so that obtained metals and inorganic matter are oxidized and denaturalized while resins are oxidized or carbonized, thereby decreasing their value; when resins contain toxic lead, separation and halogens and as materials such 20 treatment of these materials require enormous costs.

As mentioned above, for the purpose of recycling of resins, thermal decomposition of resins is not generally preferred. For this reason, there has been a proposition for a method of dissolving thermosetting resin-cured wastes in solvents, especially in organic solvents to separate inorganic matter such as metal and glass (see Japanese

20

25

Unexamined Patent Application Publication No. 10-314713).

However, because thermosetting resins including the epoxy resin are generally poorly soluble in general-purpose solvents as is clear from their definition "resin rendered insoluble and infusible by application of heat", general solvents as exemplified in the above publication do not have enough solvency for separation and recovery of resin-cured products. Therefore, pulverization is required as a pretreatment before the dissolving treatment, which places a limitation on the recycling application of recovered inorganic matter such as glass fibers in that recovered inorganic matter can be utilized only as pulverized matter and not as woven or nonwoven fabric without reprocessing.

### 15 Disclosure of Invention

It is therefore an object of the present invention to provide a treatment method involving decomposition and dissolution of an epoxy resin-cured product in order to easily recover the resin components in a recyclable state without causing thermal decomposition.

Another object of the present invention is to provide a separation method involving decomposition and dissolution of an epoxy resin-cured product to easily separate the inorganic matter and resin components contained in a composite material of inorganic matter and epoxy resin-cured product each in a recyclable state.

A further object of the present invention is to provide

10

15

20

a treatment liquid for an epoxy resin-cured product for use in the above-mentioned two methods.

In the steps of processing printed wiring boards, a variety of etching liquids have been used in order to dissolve the epoxy resin-cured product and, as an etching liquid for the epoxy resin-cured product comprising a halogenated high-molecular-weight epoxy polymer, we have reported a series of etching liquids which contain amidebased solvents and alkali metal compounds but do not contain dangerous chemicals such as concentrated sulfuric acid and chromic acid (see Japanese Unexamined Patent Application Publication No. 8-325436, No. 8-325437, No. 8-325438, No. 9-316445, and No. 10-126052). All of these etching liquids have conventionally been directed to etching and removing parts of resin-cured products to form electric circuits, and recovery and recycling of the resin component after removal have been neither meant nor intended at all. However, we have found a fact that the resin components and inorganic matter can easily be recovered in a recyclable state by using these etching liquids improved as a liquid for decomposition and dissolution of the epoxy resin-cured product.

According to one aspect of the present invention there is provided a method of treating an epoxy resin-cured product with a treatment liquid containing a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured

product. The decomposition catalyst for epoxy resin-cured products serves as a catalyst to cleave the ether linkage in the epoxy resin, thereby accelerating the decomposition of the epoxy resin swollen with the organic solvent.

According to another aspect of the present invention there is provided a method of separating a composite material of inorganic matter and epoxy resin-cured product, which comprises the following steps (1) and (2):

- (1) treating the composite material of inorganic matter 10 and epoxy resin-cured product with a treatment liquid containing a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product;
- (2) separating the inorganic matter from the liquid obtained in the step (1).

According to another aspect of the present invention there is provided a treatment liquid for an epoxy resincured product containing a decomposition catalyst for epoxy resincured products and an organic solvent.

The treatment method and separation method according to the present invention can decompose and dissolve the epoxy resin-cured product to easily recover the resin components in a recyclable state without causing thermal decomposition or without requiring pulverization of materials to be treated by using the above-mentioned treatment liquid. The decomposition products can be recycled, fur example, as a raw material for synthetic resins.

When a composite material containing inorganic matter is a subject to be treated, the inorganic matter insoluble in the treatment liquid (inorganic filler, inorganic fiber, inorganic foil, etc.) can easily be separated from the treatment liquid containing the resin components, which enables recycling of the inorganic matter as well as the resin components.

Furthermore, by applying the present invention to printed wired boards with parts mounted thereon, not only inorganic fillers and inorganic fibers, but also metal foils, which are circuits, and parts connected to the circuits are dispersed in the treatment liquid by decomposition and dissolution of the resin-cured product, thereby enabling separation and recovery of such inorganic matter.

15

20

25

10

5

Best Mode for Carrying Out the Invention

A method of treating an epoxy resin-cured product according to the present invention involves treating the epoxy resin-cured product with a treatment liquid containing a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product.

The epoxy resin-cured product to be treated is essentially comprised of an epoxy resin, a curing agent, and a crosslinking agent, and may further contain a curing accelerator, a catalyst, an elastomer, a flame retardant, etc. Components other than the epoxy resin may be present

10

as impurities.

Any epoxy resin can be an object to be treated as far as it has an epoxy group in the molecule, and examples thereof include bisphenol A-type epoxy resin, bisphenol F-type epoxy resin, bisphenol S-type epoxy resin, alicyclic epoxy resin, aliphatic linear epoxy resin, phenol novolak-type epoxy resin, cresol novolak-type epoxy resin, bisphenol A novolak-type epoxy resin, diglycidyl ethers of biphenols, diglycidyl ethers of naphthalenediols, diglycidyl ethers of phenols, diglycidyl ethers of alcohols, and compounds obtained from the above-mentioned compounds by substitution with an alkyl group, halogenation, or hydrogenation. They can be used singly or in combination.

Among the above-mentioned ероху resins, use can 15 preferably be made of epoxy resins whose hydrogen located in the ortho position on the benzene ring with respect to the ether group bonded to the same benzene group is substituted with a halogen atom such as chlorine and bromine. Such epoxy resins are obtained by reacting halogenated bisphenol 20 compounds such as tetrabromobisphenol A with epichlorohydrin, and examples include halogenated bisphenol A-type epoxy resin, halogenated bisphenol F-type epoxy resin, halogenated bisphenol S-type epoxy resin. When an electronwithdrawing halogen atom is bonded to the benzene ring of 25 the epoxy resin, cleavage of the ether linkage is more likely to occur, which can facilitate decomposition and dissolution of the epoxy resin-cured product with

10

15

25

treatment liquid.

Any curing agent for epoxy resins can be included as it can cure epoxy resins, and examples thereof include multifunctional phenols, amines, imidazole compounds, acid anhydrides, organic phosphorus compounds, and halides of the above-mentioned compounds. Any of these compounds can be used singly or in combination. Although the amount of the curing agent to be blended is not specifically limited as far as it can promote the curing reaction of the epoxy group, the curing agent is preferably included in a range of 0.01 to 5.0 equivalents, more preferably in a range of 0.8 to 1.2 equivalents, relative to one mole of the epoxy group.

the multifunctional phenols include of Examples monocyclic bifunctional phenols such as hydroquinone, resorcinol, and catechol; polycyclic bifunctional phenols such as bisphenol A, bisphenol F, naphthalenediols, and biphenols; and these compounds halogenated or substituted Novolaks and resols, which are with an alkyl group. polycondensates of these phenols and aldehydes, can be used 20 as well.

Examples of the amines include aliphatic or aromatic primary amines, aliphatic or aromatic secondary amines, aliphatic or aromatic tertiary amines, quaternary ammonium thereof, alicyclic amines, guanidines, and salts They can be, for example, any one of N,Nderivatives. benzyldimethylamine, 2-(dimethylaminomethyl)phenol, 2,4,6-

tris(dimethylaminomethyl)phenol, tetramethylguanidine, N,N'-dimethylpiperazine, triethanolamine, 1,8-diazabicyclo[5.4.0]-7diazabicyclo[2.2.2]octane, 1,5-diazabicyclo[4.4.0]-5-nonene, undecene, hexamethylenetetramine, pyridine, picoline, piperidine, 5 pyrrolidine, dimethylcyclohexylamine, dimethylhexylamine, cyclohexylamine, diisobutylamine, di-n-butylamine, diphenylamine, N-methylaniline, tri-n-propylamine, tri-ntri-n-butylamine, triphenylamine, octylamine, tetramethylammonium chloride, tetramethylammonium bromide, 10 triethylenetetramine, iodide, tetramethylammonium diaminodiphenylmethane, diaminodiphenyl ether, dicyandiamide, tolylbiguanide, guanylurea, and dimethylurea.

Examples of the imidazole compounds include imidazole, 2-ethyl-4-methylimidazole, 2-2-ethylimidazole, 15 methylimidazole, 2-phenylimidazole, 2-undecylimidazole, benzyl-2-methylimidazole, 2-heptadecylimidazole, 4,5diphenylimidazole, 2-methylimidazoline, 2-phenylimidazoline, 2-heptadecylimidazoline, 2-undecylimidazoline, isopropylimidazole, 2,4-dimethylimidazole, 2-phenyl-4-20 methylimidazole, 2-ethylidazoline, 2-phenyl-4methylimidazoline, benzimidazole, and 1-cyanoethylimidazole. acid anhydrides include phthalic the

Examples of the acid anhydrides include phthalic anhydride, hexahydrophthalic anhydride, pyromellitic dianhydride, and benzophenonetetracarboxylic dianhydride.

Any organic phosphorus compounds can be used as far as it is a phosphorus compound having an organic group.

25

Examples thereof include hexamethylphosphoric triamide, tri(dichloropropyl) phosphate, tri(chloropropyl) phosphate, triphenyl phosphite, trimethyl phosphate, phenylphosphonic acid, triphenylphosphine, tri-n-butylphosphine, and diphenylphosphine.

The epoxy resin-cured product may contain a curing accelerator. Typical examples of curing accelerators include, but are not limited to, tertiary amines, imidazoles, and quaternary ammonium salts.

The epoxy resin-cured product to be subjected to the treatment according to the present invention can be obtained by curing the epoxy resin composition comprising the above-described components using an arbitrary known method and any curing condition can be selected as far as the reaction proceeds. For example, any temperature may be used as far as the reaction preferably performed at a temperature ranging between room temperature and about 250°C. Furthermore, the curing reaction may be carried out either under pressure, under atmospheric pressure, or under reduced pressure.

Next, the treatment liquid to decompose and dissolve the above-mentioned resin-cured products contains a decomposition catalyst for epoxy resin-cured products and an organic solvent. In other words, the treatment liquid used in the present invention contains, as essential components, at least a compound that serves as a catalyst for cleavage of the ether linkage of the epoxy resin-cured products and

an organic solvent. The treatment liquid may further contain arbitrary known compounds other than the two components or even impurities.

In a preferred embodiment, the decomposition catalyst for epoxy resin-cured products contains one more 5 compounds selected from alkali metals and/or alkali metal compounds, phosphorous-containing acids and/or salts thereof, and organic acids and/or salts thereof. The term "one or more compounds" means any combination of these compounds is acceptable and examples thereof include a combination of 10 metal compounds, a combination of phosphorouscontaining acids (salts), a combination of an alkali metal compound and a phosphorous-containing acid (salt), and a combination of a phosphorous-containing acid (salt) and an organic acid (salt). These compounds are preferably added 15 in an amount of 0.001 to 80 % by weight, especially 0.1 to 30 % by weight in total in the organic solvent. When the amount is less than 0.001 % by weight, the decomposition rate of the epoxy resin-cured product tends to be lower, 용 by weight, exceeds 80 while when the amount 20 preparation of the treatment liquid tends to be difficult. Moreover, it is not always necessary that all of these compounds are dissolved in the organic solvent, and a saturated solution in which the solute is in an equilibrium state in the solution is also useful because insoluble parts 25 in the saturated solution can supply a source of fresh the already-dissolved compounds when compounds

10

15

20

25

deactivated during the reaction.

Examples of the alkali metals include lithium, sodium, potassium, rubidium, and cesium. Examples of the alkali metal compounds include hydrides, hydroxides, borohydrides, amide compounds, fluorides, chlorides, bromides, iodides, borates, phosphates, carbonates, sulfates, nitrates, organic acid salts, alcoholates, and phenolates, obtained from any one of these alkali metals. Among these compounds, the alkali metal salts are preferably used due to their high solubility in organic solvents, high catalytic effect (ion activity), and low toxicity as ions. Any of these metals and metal compounds can be used singly or in combination and preferably added in an amount of 0.01 to 80 % by weight, especially 0.1 to 10 % by weight in an organic solvent.

Examples of the phosphorous-containing acids herein include phosphoric acid, metaphosphoric acid, hypophosphoric acid, phosphorous acid (phosphonic acid), hypophosphorous acid (phosphinic acid), pyrophosphoric acid, trimetaphosphoric acid, tetrametaphosphoric acid, pyrophosphorous acid. The salts of phosphorous-containing acids used herein are salts of anions of the above-mentioned phosphorous-containing acids and cations, and the cations can be, for example, ions of lithium, sodium, potassium, rubidium, cesium, beryllium, magnesium, calcium, strontium, barium, titanium, zirconium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, silver, palladium, aluminum, gallium, stannum, or ammonium. Among these

compounds, alkali metal compounds are preferred for the same reason as described above and hydrates are preferred in terms of solubility in solvents. These salts can be any one of primary salts having a metal and two hydrogens, secondary salts having two metals and a hydrogen, and a tertiary salts having three metals. Also, these salts can be any one of acidic salts, alkaline salts, and neutral salts. Any of these compounds can be used singly or in combination and preferably added in an amount of 0.01 to 80 % by weight, especially 0.1 to 30 % by weight in an organic solvent.

Examples of the organic acids include acrylic acid, adipic acid, ascorbic acid, asparagic acid, aminobenzoic acid, alginic acid, benzoic acid, oleic acid, formic acid, citric acid, glycolic acid, gluconic acid, glutamic acid, cinnamic acid, succinic acid, acetic acid, salicylic acid, 15 oxalic acid, tartaric acid, toluenesulfonic acid, nicotinic acid, lactic acid, uric acid, halogenated acetic acid, phthalic acid, benzenesulfonic acid, malonic acid, butyric acid, and malic acid. The salts of organic acids are salts of anions of the above-mentioned organic acids and cations 20 other than hydrogen, and the cations can be, for example, lithium, sodium, potassium, rubidium, cesium, ions of beryllium, magnesium, calcium, strontium, barium, titanium, zirconium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, silver, palladium, zinc, aluminum, gallium, 25 stannum, or ammonium. Among these compounds, hydrates and alkali metal salts are preferred for the same reason as

10

15

20

described above. These salts can be any one of primary salts having a metal and two hydrogens, secondary salts having two metals and a hydrogen, and a tertiary salts having three metals. Also, these salts can be any one of acidic salts, alkaline salts, and neutral salts. Any of these compounds can be used singly or in combination and preferably added in an amount of 0.01 to 80 % by weight, especially 0.1 to 30 % by weight in an organic solvent.

As an organic solvent, one or more solvents selected from amide-based, alcohol-based, ketone-based, and etherbased solvents are preferably used in terms of solubility of ionic catalysts, but use can also be made of other solvents such as hydrocarbons, halogenated hydrocarbons, phenols, acetals, fatty acids, acid anhydrides, esters, nitrogen compounds, sulfur compounds (such as dimethylsulfoxide) and solvents having two or more functional groups (ester and ether, alcohol and ether, and the like). Any of these solvents can be used singly or in combination (for example, a combination of amide-based solvents and a combination of amide-based solvent and a non-amide-based solvent). Furthermore, any solvent can be concurrently employed and inorganic solvents such as water and ammonia may be mixed, and impurities may be present.

Preferred examples of the amide-based solvents include formamide, N-methylformamide, N,N-dimethylformamide, N,N-dimethylformamide, N,N-dimethylformamide, N,N-dimethylacetamide, N,N-di

N-methyl-2-pyrrolidone, caprolactam, and carbamates.

Examples of the alcohol-based solvents include methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, isobutanol, tert-butanol, 1-pentanol, 2-pentaol, 3-pentanol, 2methyl-1-butanol, iso-pentyl alcohol, tert-pentyl alcohol, 3-methyl-2-butanol, neopentyl alcohol, 1-hexanol, 2-methyl-1-pentanol, 4-methyl-2-pentanol, 2-ethyl-1-butanol, 1 -2-heptanol, 3-heptanol, cyclohexanol, 1 heptanol, 2-methylcyclohexanol, 3 methylcyclohexanol, methylcyclohexanol, 4-methylcyclohexanol, ethylene glycol, 10 ethylene glycol monomethyl ether, ethylene glycol monoethyl ether, ethylene glycol monopropyl ether, ethylene glycol monobutyl ether, diethylene glycol, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, diethylene glycol monopropyl ether, diethylene glycol 15 monobutyl ether, triethylene glycol, triethylene glycol monomethyl ether, triethylene glycol monoethyl ether, tetraethylene glycol, polyethylene glycol (molecular weight 1,2-propanediol, 1,3-propanediol, 400), 200 butanediol, 1,3-butanediol, 1,4-butanediol, 2,3-butanediol, 20 1,5-pentanediol, glycerol, and dipropylene glycol.

Examples of the ketone-based solvents include acetone, methyl ethyl ketone, 2-pentanone, 3-pentanone, 2-hexanone, methyl isobutyl ketone, 2-heptanone, 4-heptanone, diisobutyl ketone, cyclohexanone, methylcyclohexanone, phorone, and isophorone.

Examples of the ether-based solvents include dipropyl

10

15

20

25

ether, diisopropyl ether, dibutyl ether, dihexyl ether, anisole, phenetol, dioxane, tetrahydrofuran, acetal, ethylene glycol dimethyl ether, ethylene glycol diethyl ether, diethylene glycol dimethyl ether, and diethylene glycol diethyl ether.

Although any temperature can be used in preparing the treatment liquid, a temperature ranging between the melting point and the boiling point of the solvent used is preferred. Moreover, the treatment liquid may be prepared either in an air atmosphere or in an inert gas and either under atmospheric pressure (normal pressure), under reduced pressure, or under pressure. Other components such as surfactant may be added to the treatment liquid thus obtained.

involving decomposition method treatment The dissolution of the epoxy resin-cured product, which is a subject to be treated, with the above-mentioned treatment liquid is not specifically limited, and the treatment may be performed, for example, by immersing the epoxy resin-cured product in the treatment liquid or by spraying the treatment liquid on the epoxy resin-cured product instead of immersing. The treatment liquid may be stirred by an agitator, a pump, or blowing of gas. In the case of dipping treatment, the treatment may be conducted while vibrating the treatment The treatment liquid for the liquid by ultrasound. treatment of the resin-cured product may be used at an arbitrary temperature ranging between the freezing point and

10

15

20

25

the boiling point of the solvent in order to control the treatment rate. For the purpose of avoiding undesired thermal decomposition of the cured resin, however, the treatment liquid may preferably have a temperature of 250°C or lower in air and 300°C or lower in an inert gas. The treatment liquid may be used and stored either in air or in an inert gas, and either under atmospheric pressure, under reduced pressure, or under pressure. Although high pressure may be effective in some cases, atmospheric pressure is more preferred when safety is of concern.

size of the subject to be treated The specifically limited, and the subject may be just in the state as it is disposed of and recovered (for example, a 250 mm-square printed wiring board) or may be crushed and pulverized. A crushed and pulverized subject requires a shorter time to be treated, but the recovered materials thus obtained have limited applications. In view of this, the size of crushed pieces is preferably approximately 5 mm or larger, or 10 mm or larger. Moreover, when the crushing degree of the subject to be treated reaches pulverization (approximately 1 mm or smaller), recovered glass fibers, for example, find no application for recycling, which may significantly decrease their value. The amount of the treatment liquid with respect to the subject is specifically limited, and if the treatment liquid contacts the subject, such an amount is enough.

Methods of separating and washing, and application of

25

the resin components (the organic components derived from the resin) decomposed and dissolved in this manner are not specifically limited. For example, the resin components can be obtained by separating and removing residues (insoluble parts) from the liquid by precipitation after the treatment and removing the organic solvent by distillation. The components recovered can be recycled as a raw material for synthetic resins.

the decomposition products can be Specifically, their recovered state or preferably after 10 recycled in purification, when the decomposition products obtained after the treatment of the resin-cured product are, for example, phenols, glycidyl ethers of phenols, metal salts of phenols, amines, carboxylic acids, and halogenated or hydrogenated products of the above-mentioned compounds (in the concrete, 15 cresol, dimethylphenol, propylphenol, phenol, such ethylphenol, hydroquinone, resorcinol, catechol, bisphenol A, biphenol, dihydroxydiphenyl F, ether, bisphenol dihydroxydiphenyl sulfone, phenol novolak, cresol novolak, bisphenol A novolak, and glycidyl ethers of these compounds, 20 halides of these compounds, alkali metal salts of these compounds, and ammonium salts of these compounds).

Next, a method of separating a composite material of inorganic matter and epoxy resin-cured product according to the present invention includes the following steps (1) and (2);

(1) treating the composite material of inorganic matter

and epoxy resin-cured product with a treatment liquid containing a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product; and

(2) separating the inorganic matter from the liquid obtained in the step (1).

5

10

Composite materials of inorganic matter and epoxy resin-cured product to be treated comprise the above-mentioned epoxy resin-cured product and inorganic matter, and are typically insulating boards, metal-clad laminates, or printed wiring boards, which contain metals and glass as inorganic matter.

The inorganic matter can be, for example, metals and metal oxides, hydroxides, halides, or nitrides. concretely, examples include boron, aluminum, iron, silicon, 15 titanium, chrome, cobalt, nickel, zinc, palladium, silver, stannum, tungsten, platinum, gold, lead, alumina, zirconia, titania, magnesia, silicon carbide, silicon nitride, boron nitride, mica, silica, clay, glass, carbon, calcium carbonate, aluminum hydroxide, magnesium hydroxide, 20 These metals and compounds may be fused calcium silicate. The shape of the inorganic matter may be, for or mixed. example, powder, granule, fiber, foil, film, wire, The fiber may be in a mat-like state or woven like circuit. Although the content of inorganic matter in a 25 fabric. composite material is arbitrary, it generally ranges from 5 to 90 % by weight based on the total amount of the composite material.

When the inorganic matter is, for example, powdery, granular, or staple-like, such inorganic matter is blended with the above-mentioned epoxy resin composition for curing, 5 which is cured by cast molding or the like to obtain a composite material of inorganic matter and epoxy resin-cured product. When the inorganic matter is mat-like or fabricsuch inorganic matter is immersed in the abovementioned epoxy resin composition (which may contain powdery, 10 granular, or staple-like inorganic matter as above) and then partial-cured to obtain a B-stage prepreg. Such a B-stage prepreg and laminates obtained by laminating and curing this prepreg or a prepreg at a pre-B-stage are also the composite material of inorganic matter and epoxy 15 resin-cured product. The present invention is further applicable to the composite materials of inorganic matter and epoxy resin-cured product such as: resin boards or films covered with a metal foil obtained by coating or flowcoating the metal foil with the above-mentioned epoxy resin 20 composition (which may contain powdery, granular, or staplelike inorganic matter as described above) and then curing the same; resin boards covered with a metal foil obtained by laminating a metal foil onto the above-mentioned prepred or prepreg at a pre-B-stage and then curing the same; laminates 25 covered with a metal foil obtained by laminating the abovementioned metal foil-covered resin boards or films (before curing) or metal foil-covered resin boards (which are formed

with prepregs and before curing) onto the above-mentioned laminates and then curing the same; interlayer circuit boards obtained by using the above-mentioned epoxy resin composition (which may contain powdery, granular, or staplelike inorganic matter as described above) and metal foils and applying an appropriate circuit formation method; and laminates covered with a metal foil obtained by laminating the above-mentioned metal foil covered-resin boards or films (before curing) or the metal foil covered-resin boards (which are formed with prepregs and before curing) onto the above-mentioned interlayer circuit boards and then curing The above-mentioned resin boards, films, or the same. laminates, which have metal foils, may be ones having a circuit formed by applying a circuit formation method to metal foils.

5

10

15

20

25

The step (1) of treating the composite materials, the resin components thus obtained and recycling thereof are the same as described for the treatment method of the epoxy separation step (2) is not resin-cured product. The specifically limited and inorganic matter such as metals and glass can easily be separated from the liquid, for example, recycling decantation, for purpose. filtration or According to the separation method of the present invention, because subjects to be treated such as substrate need not to be crushed or pulverized before the treatment, inorganic matter can be recovered in such a recyclable state that is suitable for a variety of applications. For example, it is possible to recycle glass fibers (glass fabrics) in their recovered state.

Furthermore, a treatment liquid for an epoxy resincured product according to the present invention contains a decomposition catalyst for epoxy resin-cured products and an 5 organic solvent and is preferably used in the method of treating the epoxy resin-cured product according to the present invention and in the method of separating composite materials of inorganic matter and epoxy resin-cured product The preferred the present invention. according to 10 components and preparation method of the treatment liquid are the same as described for the above-mentioned method of treating the epoxy resin-cured product.

## 15 EXAMPLES

The present invention is illustrated in detail with reference to the examples, which are not intended to limit the present invention. In the following examples, % means % by weight.

20

25

A. Solubility in treatment liquid containing alkali metal compound (1)

[EXAMPLES A1 to A15 and COMPARATIVE EXAMPLES A1 to A6]
(Preparation of epoxy resin composition)

Epoxy resin compositions were prepared by blending a brominated bisphenol A-type epoxy resin (manufactured by Sumitomo Chemical, Co., Ltd., product name "ESB400T"; epoxy

10

15

20

25

equivalent 400, bromine content 48%), a bisphenol A-type epoxy resin (manufactured by Yuka Shell Epoxy Kabushiki Kaisha, product name "Epikote 1001"; epoxy equivalent 470), a phenol novolak resin (manufactured by Hitachi Chemical Co., Ltd., product name "HP850N"; hydroxyl equivalent 106) as a curing agent, and imidazole as a curing accelerator. The amounts of the two epoxy resins and the phenol novolak resin were adjusted such that the ratio of the epoxy equivalent to the hydroxyl equivalent was 1. The amount of each epoxy resin was adjusted such that two samples of the epoxy resin composition with a respective bromine content of 20% and 30% relative to the total amount of the epoxy resin composition were obtained. The curing agent was added in an amount of 0.5% based on the total amount of the epoxy resins.

Two samples of the epoxy resin composition obtained in this manner were cured in an oven at 170°C for 60 minutes to obtain two kinds of resin boards with different bromine contents (about 0.5 to about 1.0 mm thick).

(Preparation of treatment liquid)

In each case of Examples Al to Al5, components listed in Table 1 were mixed to prepare a 3% solution of an alkali metal compound in an amide-based, ketone-based, alcoholbased or ether-based organic solvent. In some of these treatment liquids, the alkali metal compound was not completely dissolved and precipitated on the bottom of a tank when the liquids were allowed to stand.

The abbreviations for the organic solvents in Table 1

are as follows: NMP, N-methyl-2-pyrrolidone; CHON, cyclohexanone; PEG, polyethylene glycol #200; DGMM, diethylene glycol monomethyl ether; and DGDM, diethylene glycol dimethyl ether.

5 Treatment liquid that contained no alkali metal compound was used in each case of Comparative Examples A1 to A5, and a 3% aqueous solution of sodium hydroxide was prepared in Comparative Example A6.

(Treatment of epoxy resin-cured product)

The two resin boards were cut into pieces of a size of about 5 mm square each and 0.5 g of the pieces were weighed out and placed in 50 g of each treatment liquid. The treatment liquids were maintained at a predetermined temperature (60°C, 80°C, or 100°C) and agitated hard for one hour. The treatment liquids were visually inspected to evaluate the solubility according to the following rating. 5: all dissolved; 4: almost all dissolved; 3: half dissolved; 2: significantly broken; 1: broken to some extent; 0: no change.

The results are shown in Table 1.

[Table 1] A. Solubility in treatment liquid containing alkali metal compound (1)

A   Treatment   CO & by weight   SO & by weight     A   NNP	Example	Compos	Composition of	Brom	Bromine content	nt:	Brom	Bromine content	ent:
Naclarid   60°C   80°C   100°C   60°C   80°C   100°C     NMP   4   5   5   5   5   5     CHON   2   3   4   4   4   5     DCDM   1   2   3   3   4   5     DCDM   0   1   3   3   4   5   5     DCDM   0   1   3   3   4   5   5     DCDM   0   1   2   3   4   5   5     DCDM   0   1   2   2   3   3   4   5     DCDM   0   1   2   2   3   3   3     DCDM   0   1   3   5   3   5   5     Na2Co   PEG   0   0   1   1   2   2   3   3     DCDM   0   1   2   2   3   3   3     DCDM   0   0   0   0   0   0     CHON   0   0   0   0   0   0     DCDM   0   0   0   0   0     DCDM   0   0   0   0   0     DCDM   0   0   0   0   0     KOH   Water   0   0   0   0   0     DCDM   Water   0   0   0   0     TOTAL   DCDM   0   0   0   0     TOTAL   DCDM   0   0   0   0     TOTAL   DCDM   DCDM   DCDM   DCDM   DCDM   DCDM     TOTAL   DCDM		trea	tment	20	%	ht	30	%	
KOH         PEG         0         1         2         5         5         5           CHON         2         3         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4<		11¢	luid		၁ 08			၁့ 08	100 °C
KOH         PEG         0         1         2         3         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         4         5         3         3         4         5         3         4         5         3         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         6         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7 <td>A 1</td> <td></td> <td>NMP</td> <td>4</td> <td>2</td> <td>2</td> <td>5</td> <td>5</td> <td>5</td>	A 1		NMP	4	2	2	5	5	5
KOH         PEG         0         1         2         3         3         4         1         2         3         4         4         1         2         3         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         4         5         6         7         6         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7 <td>A 2</td> <td></td> <td>CHON</td> <td>2</td> <td>3</td> <td>4</td> <td>4</td> <td>4</td> <td>5</td>	A 2		CHON	2	3	4	4	4	5
Nacl   DGMM   1   1   2   3   4	A 3	КОН	PEG	0	1	2	3	3	5
Nacl DGDM 0 1 3 3 4 5 4 5 CHON 1 2 3 2 4 5 CHON 1 2 3 2 4 5 CHON 1 2 3 2 4 5 CHON 0 1 1 2 2 2 2 CHON 0 1 1 3 5 3 5 CHON 0 0 1 1 1 2 CHON 0 0 1 1 1 2 CHON 0 0 0 0 0 0 0 0 0 0 0 CHON 0 0 0 0 0 0 CHON 0 0 0 0 0 CHON 0 0 0 0 0 0 CHON 0 0 CHON 0 0 0 CHON 0 CHON 0 0 CHON 0 C	A 4		DGMM	-	1	2	3	4	5
NaCl PEG 0 0 1 1 2 3 4 5 4 5 4 5 6 4 5 6 4 6 6 6 6 6 6 6 6 6	A 5		DGDM	0	1	3	3	4	5
NaCl PEG         PEG         0         0         1         2         4           DGMM         0         1         1         2         2           DGDM         0         1         3         2         3         2           DGDM         0         1         3         2         3         5         3         5           CHON         0         2         2         2         3         5         3         5         1         2         3         6         1         2         3         6         1         2         3         6         1         2         3         6         1         2         3         6         1         2         3         6         1         2         3         6         1         2         3         6         1         2         3         6         3         6         1         1         2         3         6         3         6         1         1         2         3         7         3         6         1         1         2         3         1         1         2         3         3         1         1         1	A 6		NMP	3	4	2	4	2	2
NaCl         PEG         0         0         1         1         2           DGMM         0         1         2         2         2           DGDM         0         1         3         5         3         5           CHON         0         2         2         3         5         3         5           CHON         0         0         1         3         5         3         5         1           DGMM         0         0         1         2         2         3         5         1           DGDM         0         1         2         2         3         5         1           DGDM         0         1         2         2         3         5         1           CHON         0         1         2         2         3         5         1           NMP         0         0         0         0         0         0         0         0         0           -         PEG         0         0         0         0         0         0         0         0         0         0         0         0         0	A 7		CHON	-	2	ဆ	2	4	5
Ma2CO3 DGMM 0 1 2 2 2 3	A 8	NaCl	PEG	0	0	-	1	2	2
NAMP 1 3 5 3 5 7 7 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	A 9		DGMM	0	1	2	2	2	က
Na <sub>2</sub> CO <sub>3</sub> PEG 0 0 1 1 2 3 5 5 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7	A 10		DGDM	0	1	3	2	3	က
Na <sub>2</sub> CO <sub>3</sub> PEG         0         2         2         3         8           DGMM         0         1         1         2         3         8           .         DGDM         0         1         2         2         3         8           .         DGDM         0         1         2         2         3         8           .         NMP         0         0         0         0         0         0         0           .         THON         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0 </td <td>A 11</td> <td></td> <td>NMP</td> <td>_</td> <td>က</td> <td>2</td> <td>3</td> <td>2</td> <td>5</td>	A 11		NMP	_	က	2	3	2	5
Na <sub>2</sub> CO <sub>3</sub> PEG         0         0         1         1         2         2           DGMM         0         1         2         2         3         1           .         DGDM         0         1         2         2         3         1           .         NMP         0         0         0         0         0         0         0           .         PEG         0         0         0         0         0         0         0         0           .         PEG         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0 <td>A 12</td> <td></td> <td>CHON</td> <td>0</td> <td>2</td> <td>2</td> <td>2</td> <td>သ</td> <td>4</td>	A 12		CHON	0	2	2	2	သ	4
- DGMM 0 1 2 2 3 8 PEG 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	A 13	Na2CO3	PEG	0	0	1	1	2	2
. DGDM 0 1 2 2 3 6	A 14		DGMM	0	,	2	2	က	က
. NMP 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	A 15		DGDM	0	1	2	2	က	3
- PEG 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	Comp. Ex.								
- PEG 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	A 1		NMP	0	0	0	0	0	0
-         PEG         0         0         0         0         0         0         0           DGMM         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0         0 <td>A 2</td> <td></td> <td>CHON</td> <td>0</td> <td>0</td> <td>0</td> <td>0</td> <td>0</td> <td>0</td>	A 2		CHON	0	0	0	0	0	0
KOH         Water         0         0         0         0         0         0           KOH         Water         0         0         0         0         0         0	A 3	ì	PEG	0	0	0	0	0	0
KOH         Water         0         0         0         0         0         0	A 4		DGMM	0	0	0	0	0	0
KOH Water 0 0 0 0 0 0	A 5		DGDM	0	0	0	0	0	0
	A 6	КОН	Water	0	0	0	0	0	0

10

20

As shown in Table 1, when the resin boards were treated with an organic solvent alone as in Comparative Examples Al to A5, the resin-cured products were not dissolved. In Comparative Example A6, where the resin boards were treated with an aqueous solution of an alkali metal compound, the result was the same.

In contrast, in Examples A1 to A15, when the resin boards were treated with an organic solvent solution of an alkali metal compound, solubility was good in all the cases and the resin was dissolved completely in some cases.

B. Solubility in treatment liquid containing alkali metal compound (2)

[EXAMPLES B1 to B15, COMPARATIVE EXAMPLES B1 to B6]

- 15 (Preparation of composite material of inorganic matter and epoxy resin-cured product)
  - (1) The following two kinds of copper-clad laminate were provided: "MCL-E-679" (bromine content in the resincured product is 15%) and "MCL-E-67" (bromine content in the resin-cured product is 20%); both which are manufactured from a brominated epoxy resin, a bromine-free epoxy resin, a curing agent, a curing accelerator, glass cloth, and copper foil, and both are product names of Hitachi Chemical Co., Ltd.
- 25 (2) An epoxy resin composition was prepared by blending a brominated bisphenol A-type epoxy resin (manufactured by Tohto Kasei Co., Ltd., product name "AER8011"; epoxy

10

15

20

equivalent 470, bromine content 20%), a high-molecularweight brominated bisphenol A-type epoxy resin (bromine content 53%, number average molecular weight 25,000; the number average molecular weight was measured by gel permeation chromatography and converted based calibration curve prepared with a standard polystyrene), a phenol novolak resin (manufactured by Hitachi Chemical Co., Ltd., product name "HP850N"; hydroxyl equivalent 106) as a curing agent, and 2-methyl-4-methylimidazole as a curing accelerator. The amounts of the two epoxy resins and the phenol novolak resin were adjusted such that the ratio of the epoxy equivalent to the hydroxyl equivalent was 1. amount of each epoxy resin was adjusted to give a bromine content of 28% relative to the total amount of the epoxy resin composition excluding the amount of inorganic matter. The curing agent was added in an amount of 0.5% based on the total amount of the epoxy resins.

Glass cloth was impregnated with the epoxy resin composition obtained and predried at 160°C for 4 minutes to form a prepreg. A copper foil was laminated onto the prepreg, which was then dried at 170°C for 90 minutes to obtain a copper-clad laminate, which is a composite material of inorganic matter and epoxy resin-cured product. This is referred to as "self-made laminate sample" hereinafter.

25 (3) Two kinds of the copper-clad laminate of (1) and the self-made laminate sample of (2) were cut into pieces of a size of 10 mm by 50 mm each and three lines of circuit of

10

20

25

1 mm-wide copper foils were formed on the both sides of each piece to obtain test pieces.

(Preparation of treatment liquid)

In each case of Examples B1 to B15, components listed in Table 2 were mixed to prepare a 3% solution of an alkali metal compound in an amide-based, ketone-based, alcoholbased or ether-based organic solvent(s). The combinations shown in Examples B1, B2, and B5 were NMP 80% and PEG 17%, and DGMM 17%, and DGDM 80% and 808 CHON In some of these treatment liquids, respectively. alkali metal compound was not completely dissolved and precipitated on the bottom of a tank when the liquids were allowed to stand. The compound names in Table 2 are the same as in Table 1.

Treatment liquid that contained no alkali metal compound was used in each case of Comparative Examples B1 to B5, and a 3% aqueous solution of sodium hydroxide was prepared in Comparative Example B6.

(Treatment of composite material)

The above-mentioned three kinds of test pieces were weighed and then immersed in each of the treatment liquids at a predetermined temperature (60°C or 100°C). After 60 minutes, the test pieces were taken out from the treatment liquid and weighed again. The weight of the resin-cured product before the treatment was calculated from the ratio of the resin in the composite material, and the rate of change in weight of the resin-cured product was calculated

from the difference in weight before and after the treatment to obtain a solubility of the resin-cured product in the treatment liquid.

The results are shown in Table 2.

(Solubility /%) [Table 2] B. Solubility in treatment liquid containing alkali metal compound (2)

Example	Сошро	osition of	Tempe	Temperature:	၁ 09	Тешре	Temperature:	: 100 °C
4	tre	reatment	E-679	1	Sample*	E-679		Sample*
		liguid	Br 15wt%	Br 20wt%	Br 28wt%	Br 15wt%	Br 20wt%	<b>Br 28wt%</b>
B 1		NMP, PEG	1.3	6.4	48.0	2.5	L.62	93.5
B 2		CHON, DGMM	2.1	2.2	12.3	5.9	5.4	57.8
B 3	КОН	9aa	2.2	2.3	3.3	2.3	6.1	33.7
B 4		DGMM	1.7	2.1	9.6	2.6	16.3	89.0
B 5		DGDM, PEG	1.8	2.1	12.7	2.9	5.4	57.8
B 6		AWN	6.0	3.5	25.1	3.2	4.5	68.0
B 7		CHON	1.9	1.7	11.6	2.0	2.4	23.8
B 8	NaCl	PEG	2.1	1.8	3.4	2.7	2.1	4.2
B 9		DGMM	1.7	1.2	7.2	2.3	1.8	8.3
B 10		DGDM	1.5	1.8	9.4	1.6	2.8	9.8
B 11		AWN	0.9	1.8	25.4	1.9	23.4	53.3
B 12		CHON	1.8	1.7	9.7	1.9	3.7	22.6
B 13	Na <sub>2</sub> CO <sub>3</sub>	PEG	2.1	1.5	3.3	1.7	1.9	3.1
B 14		DGMM	2.0	2.7	9.6	2.3	3.2	10.0
B 15		DGDM	2.2	1.6	13.4	2.0	3.0	13.8
Comp. Ex.								
B 1		NMP	0.4	0.2	9.0	0.8	0.5	0.9
B 2		CHON	0.3	0.3	0.4	ı	1	ı
В 3	1	PEG	0.2	0.2	0.3	1	1	1
B 4		DGMM	. 0.4	0.3	0.5	1	l	ı
B 5		DGDM	0.5	0.4	0.4	1	1	ı
B 6	КОН	Water	0.3	0.1	0.2	0.2	0.4	0.8

\* self- made laminate sample

10

20

As shown in Table 2, when treated with an organic solvent alone as in Comparative Examples B1 to B5, no test piece showed a solubility exceeding 1%. Also, even when treated with an aqueous solution of an alkali metal compound as in Comparative Example B6, the solubility was below 1%.

In contrast, when the treatment liquid was an organic solvent solution of an alkali metal compound as in Examples B1 to B15, a maximum solubility of 93.5% was obtained. Although the composite material with lower bromine content showed a lower solubility, the solubility was two to ten times as high as that in Comparative Examples.

- C. Solubility in treatment liquid containing a phosphorouscontaining acid/ its salt
- 15 [EXAMPLES C1 to C45, COMPARATIVE EXAMPLES C1 to C21]

  (Preparation of composite material of inorganic matter and epoxy resin-cured product)

A brominated epoxy resin-cured product, glass cloth, and copper foil were used to form a composite material, which was heated to 170°C for 90 minutes to cure. Dicyandiamide, which is an amine, was added as a curing agent of the brominated epoxy resin. The amount of the brominated epoxy resin was adjusted to yield a bromine content of the resin-cured product of approximately 20%.

25 The sample was cut into a size of 10 mm by 30 mm and three lines of circuit of 1 mm-wide copper foils were formed on the both sides of the cut sample to obtain a test piece.

10

20

25

(Preparation of treatment liquid)

In each case of Examples C1 to C45, a phosphorous-containing acid or its salt listed in Table 3 was weighed in an amount corresponding to 1.0 equivalent with respect to one liter of solvent and mixed with the solvent at room temperature to prepare a treatment liquid. The compound names of the organic solvents shown in Table 3 are the same as in Examples A.

Comparative Examples are: Comparative Examples C1 to C3 where each treatment liquid contained an organic solvent only; Comparative Examples C4 where the treatment liquid contained water only; and Comparative Examples C5 to C21 where each treatment liquid was a 3% aqueous solution of a phosphorous-containing acid or its salt listed in Table 3.

15 (Preparation of composite material)

Each of above-mentioned treatment liquids was placed in a flask equipped with a condenser, a thermometer, an inlet for nitrogen, and a stirrer and heated to 140°C (for organic solvents) or 100°C (for aqueous solutions) in an oil bath while being mildly stirred under nitrogen gas stream. The above-mentioned test piece was weighed, immersed in each treatment liquid for four hours, taken out and weighed again. The change in weight before and after the treatment was divided by the total weight of the resin in the test piece which was weighted beforehand to obtain a solubility of the resin-cured product.

The results are shown in Table 3.

(Solubility /%) [Table 3] C. Solubility in treatment liquid containing phosphorous-containing acid /its salt

<u>.</u>																									
7777	<b>%</b>		0.8	1.2	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0		
A TTTONTOC)	Conposition		NMP	DGMM	CHON	W (=Water)	HPO3 + W	H3PO4 + W	H <sub>3</sub> PO <sub>3</sub> + W	Na3PO4.12H2O + W	(NaPO3) n + W	Na2HPO4 + W	Na2HPO4.12H2O + W	NaH2PO4.2H2O + W	Na2HPO3.5H2O + W	NaPH2O2.H2O + W	K3PO4.nH2O + W	$(KPO_3)_n + W$	Mg3PO4·8H2O + W	AlPO4 + W	Ca3(PO4)2 + W	FePO4.nH2O + W	(NH4) 3PO4 · 3H2O + W		
	Comp.	EX.	C 1	C 2	င ၁	C 4	C 2	9 O	C 7	C 8	6 0	C 10	C 11	C 12	C 13	C 14	C 15	C 16	C 17	C 18	C 19	C 20	C 21		
	%		6.5	2.2	0.8	11.0	1.6	6.0	20.5	33.7	6.4	3.8	1.1	0.2	7.4	1.5	2.2	1.8	1.8	1.1	3.7	1.2	5.8	1.6	
	ion		NMP	DGMM	CHON	NMP	DGMM	CHON	NMP	DGMM	CHON	NMP	DGMM	CHON	NMP	DGMM	NMP	DGMM	NMP	DGMM	NMP	DGMM	NMP	DGMM	
	Composition		Na <sub>2</sub> HPO <sub>3</sub>	.5H2O		NaPH2O2	.H2O		K3PO4	nH2O			(KPO3) n		Mg3PO4	02H8.	A1PO4		Ca3(PO4)2		FeP04	OZHu.	(NH4) 3PO4	·3H20	
	Ex.		C 24	C 25	C 26	C 27	C 28	C 29	C 30	C 31	C 32	C 33	C 34	C 35	C 36	C 37	C 38	C 39	C 40	C 41	C 42	C 43	C 44	C 45	
	96/		3.6	1.2	0.4	13.9	0.3	0.1	11.4	0.2	0.1	37.6	28.1	4.5	2.1	١.	• •	2.5	2.3	0.3	6.1	1.0	13.0	1.2	0.4
	tion		NMP	DGMM	CHON	NMP	DGMM	CHON	NMP	DGMM	CHON	NMP	DGMM	CHON	NMP	DGMIM	CHON	NMP	DGMM	CHON	NMP	CHON	NMP	DGMM	CHON
	Composition			HPO3	<del></del>		H3P04	· ·		H3PO3		Na <sub>3</sub> PO <sub>4</sub>	·12H2O			n(soden)			Na2HPO4		Na2HPO4	·12H20	NaH2P04	·2H20	
	Ex.		C 1	C 2	C 3	C 4	C 5	90	C 2	80	6 ට	C 10	C 11	C 12	C 13	C 14	C 15	C 16	C 17	C 18	C 19	C 20	C 21	C 22	C 23

10

15

20

As shown in Table 3, the solubility was approximately 1% in Comparative Examples C1 to C3 where the treatment liquid was only an organic solvent. And the solubility was 0% in Comparative Examples C4 to C21 where the treatment liquid was only water or an aqueous solution of a phosphorous-containing acid or its salt.

In contrast, the treatment liquid dissolved the resincured product in all the cases of Examples C1 to C45 where the treatment liquid was an organic solvent solution of a phosphorous-containing acid or its salt although the solubility varied depending on the solvent. In particular, high solubility was shown in Examples C10, C11, C30, and C31.

D. Solubility in treatment liquid containing organic acid or its salt

[EXAMPLES D1 to D34, COMPARATIVE EXAMPLES D1 to D20]

A composite material of inorganic matter and epoxy resin-cured product was prepared as in Examples C and subjected to the same treatment as in Examples C with a respective one of the treatment liquids containing the components listed in Table 4 to determine the solubility in each treatment liquid.

The results are shown in Table 4.

[Table 4] D. Solubility in treatment liquid containing organic acid/its salt

[rable 4]	[] D. Solubility in treatment liquid	ent lic	luid co	containing	g organic acid/its salt (Solubility	
Ex.	Composition		%	Comp. Ex.	Composition	d10
D1	Lithium acetate	NMP	8.2	D1	NMP	0.8
102		DGMM	2.1	D2	DGMM	•
D3	Lithium acetate	NMP	21.3	D3	W (=Water)	•
D4	dihydrate	DGMM	3.2	D4	Lithium acetate + W	•
D5	Lithium citrate	NMP	١ •	DS	<b>≥</b> +	•
D6	tetrahydrate	DGMIM	12.3	9Q	Z	-
D7	Sodium acetate	NMP	7.4	D7	+	•
D8		DGMM	``	D8	zoate + W	•1
D9	Sodium benzoate	NMP	12.5	60	3	- 1
D10		DGMM	1.3	D10	<u>nexahydrat</u>	0.0
D11	Sodium citrate	NMP	7.0	D11		•1
D12	dehydrate	DGMM	2.1	D12	1fonate	0.0
D13	Sodium succinate	NMP	9.3	D13	Potassium acetate + W	0.0
D14	hexahydrate	DGMM	2.0	D14	Potassium benzoate + W	0.0
D15	Sodium tartrate	NMP	6.5	D15		0.0
D16	dihydrate	DGMM	2.4	D16		0.0
D17	Sodium benzenesulfonate	NMP	1.9	D17	Calcium acetate hydrate + W	0.0
D18	hydrate	DGMM	1.3	D18	Iron citrate hydrate + W	0.0
D19	Potassium acetate	NMP	22.0	D19	Ammonium acetate + W	0.0
D20		DGMM	13.3	D20	Tartaric acid + W	0.0
D21	Potassium benzoate	NMP	10.2			
D22		DGMM	1.6	· ·		
D23	Potassium citrate	NMP	•			
D24		DGMM	•			
D25	Aluminium acetate	NMP	3.5			
D26	hydrate	DGMM	1.7			
D27	Calcium acetate	NMP	7.3			
D28	hydrate	DGMM	1.6			
D29	Iron citrate	NMP	3.3			
D30	hydrate	DGMM	2.1			
D31	Ammonium acetate	NMP	6.4			
D32		DGMIM	1.4			
D33	Tartaric acid	NMP	1.3	<del></del>		
D34		DGMM	1.9			

As shown in Table 4, the solubility was approximately 1% in Comparative Examples D1 and D2 where the treatment liquid was an organic solvent. The solubility was 0% in Comparative Examples D3 to D20 where the treatment liquid was only water or an aqueous solution of an organic acid or its salt.

In contrast, the treatment liquid dissolved the resincured product in all the cases of Examples D1 to D34 where the treatment liquid was an organic solvent solution of an organic acid or its salt, although the solubility varied depending on the solvent. In particular, high solubility was shown in Examples D3, D6, D9, D19, D20, and D21.

The present disclosure relates to subject matter contained in Japanese Patent Applications NO. HEI11-286187, filed on October 7, 1999, and No. HEI11-286188, filed on October 7, 1999, the disclosure of which is incorporated herein by reference.

It is to be noted that, besides those already mentioned above, many modifications and variations of the above embodiments may be made without departing from the novel and advantageous features of the present invention. Accordingly, all such modifications and variations are intended to be included within the scope of the appended claims.

5

10

10

15

20

## CLAIMS

- 1. A method of treating an epoxy resin-cured product with a treatment liquid containing a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product.
- 2. The method according to claim 1, wherein the decomposition catalyst for epoxy resin-cured products contains one or more compounds selected from alkali metals and/or alkali metal compounds, phosphorous-containing acids and/or salts thereof, and organic salts and/or salts thereof.
- 3. The method according to claim 1, wherein the organic solvent contains one or more solvents selected from amidebased solvents, alcohol-based solvents, ketone-based solvents, and ether-based solvents.
- 4. The method according to claim 2, wherein the alkali metal compounds are alkali metal salts.
- 5. The method according to claim 2, wherein the salts of phosphorous-containing acids are hydrates and/or alkali metal salts.
- 6. The method according to claim 2, wherein the salts of organic acids are hydrates and/or alkali metal salts.
- 7. The method according to claim 1, wherein the epoxy resin-cured product contains a halogen atom.
- 1, wherein a claim method according to 8. The 25 decomposition product of the epoxy resin-cured product contains a compound recyclable as material of a raw

20

25

synthetic resins.

- 9. The method according to claim 1, wherein the epoxy resin-cured product is treated under atmospheric pressure.
- 10. The method according to claim 1, wherein the treatment liquid has a temperature of 250°C or lower in air and 300°C or lower in an inert gas.
  - 11. A method of separating a composite material of inorganic matter and epoxy resin-cured product, comprising the steps of:
- 10 (1) treating the composite material of inorganic matter and epoxy resin-cured product with a treatment liquid containing a decomposition catalyst for epoxy resin-cured products and an organic solvent to decompose and dissolve the epoxy resin-cured product; and
- 15 (2) separating the inorganic matter from the liquid obtained in the step (1).
  - 12. The method according to claim 11, wherein the decomposition catalyst for epoxy resin-cured products contains one or more compounds selected from alkali metals and/or alkali metal compounds, phosphorous-containing acids and/or salts thereof, and organic salts and/or salts thereof.
  - 13. The method according to claim 11, wherein the organic solvent contains one or more solvents selected from amidebased solvents, alcohol-based solvents, ketone-based solvents, and ether-based solvents.
  - 14. The method according to claim 12, wherein the alkali metal compounds are alkali metal salts.

WO 01/25317 PCT/JP00/06951

15. The method according to claim 12, wherein the salts of phosphorous-containing acids are hydrates and/or alkali metal salts.

- 39 -

16. The method according to claim 12, wherein the salts of organic acids are hydrates and/or alkali metal salts.

5

20

- 17. The method according to claim 11, wherein the epoxy resin-cured product contains a halogen atom.
- 18. The method according to claim 11, wherein the inorganic matter is metal and/or glass.
- 10 19. The method according to claim 11, wherein the composite material of inorganic matter and epoxy resin-cured product is any one of an insulating board, a metal-clad laminate, and a printed wiring board.
- 20. The method according to claim 11, wherein a decomposition product of the epoxy resin-cured product contains a compound recyclable as a raw material of synthetic resins.
  - 21. The method according to claim 11, wherein the epoxy resin-cured product is treated under atmospheric pressure in the step (1).
  - 22. The method according to claim 11, wherein the treatment liquid in the step (1) has a temperature of 250°C or lower in air and 300°C or lower in an inert gas.
- 23. A treatment liquid for an epoxy resin-cured product
  25 containing a decomposition catalyst for epoxy resin-cured products and an organic solvent.
  - 24. The treatment liquid according to claim 23, wherein the

decomposition catalyst for epoxy resin-cured products contains one or more compounds selected from alkali metals and/or alkali metal compounds, phosphorous-containing acids and/or salts thereof, and organic salts and/or salts thereof.

25. The treatment liquid according to claim 23, wherein the organic solvent contains one or more solvents selected from amide-based solvents, alcohol-based solvents, ketone-based solvents, and ether-based solvents.